

## Evaluation of Magnetostriction of the Single-Variant Ni–Mn–Ga Martensite

Victor A. L'vov<sup>1,a</sup>, Ilya Glavatsky<sup>2,b</sup> and Nadiya Glavatska<sup>2,c</sup>

<sup>1</sup>Department of Radiophysics, Taras Shevchenko University, 01601, Kyiv, Ukraine

<sup>2</sup>Institute for Metal Physics, NAS of Ukraine, 03142, Kyiv, Ukraine

<sup>a</sup>victorlvov@univ.kiev.ua, <sup>b</sup>glavat@imp.kiev.ua, <sup>c</sup>lglav@imp.kiev.ua

**Keywords:** Ferromagnetic martensite. Elastic modulus. Magnetostriction.

**Abstract.** The temperature dependence of ordinary magnetostriction of the axially compressed Ni–Mn–Ga alloy with the low values of shear elastic modulus  $C(T) \sim 1 - 10$  GPa has been evaluated theoretically in the framework of Landau theory. The computations showed that the compression with 50 MPa stress reduces the ordinary magnetostriction by factor 3 at room temperature. Nevertheless, the magnetostriction of compressed alloy exceeds the value of  $10^{-4}$  in the whole temperature range of martensitic phase stability, strongly depends on the temperature in the vicinity of martensitic transformation (MT), and is practically constant well below MT temperature. Therefore, the purposeful search for the alloy with the low value of shear elastic modulus and high MT temperature (well above 300 K) may result in the discovery of good magnetostrictive material. This material will possess the temperature-independent magnetostriction value about of  $10^{-4} - 10^{-3}$  and rather low electric conductivity enabling the technical applications of this material in dynamic regimes.

### Introduction

The most intensively studied ferromagnetic shape memory alloys (FSMA-s) belong to the Ni–Mn–Ga alloy family (see e.g. review article [1]). These alloys exhibit the giant magnetically induced deformation [2] in combination with the large ordinary magnetostriction and ultra-low values of shear elastic modulus. However, the considerably different magnitudes of ordinary magnetostriction (from  $10^{-5}$  to  $10^{-3}$  [3–6]) and shear modulus (from 1 GPa to 60 GPa [7–10]) have been observed by the different authors. The existing experimental results are disputed and the tendencies to reduction of the reported values of magnetostriction (on the one hand) and shear modulus (on the other hand) take place. These tendencies are contradictory, because the magnetostriction is inversely proportional to the elastic stiffness of the alloy specimen.

The difficulties in the measurements of elastic modules and magnetostriction are mainly caused by the easy transformable martensitic structure of FSMA-s. The structural transformation of martensite under the test load or magnetic field is accompanied by the deformation of alloy specimen, which exceeds the elastic deformation or ordinary magnetostriction and retards the comprehension of experimental results. It was observed, however, that the compressive mechanical stress of 5 Mpa is sufficient for the blocking of magnetic field action on the martensitic structure [2,11]. Hence, the magnetostriction measurements under the permanent mechanical load became of particular interest [6].

In the present article the magnetostriction of axially compressed ultra-soft Ni–Mn–Ga alloy with the temperature-dependent shear elastic modulus varying from 1 GPa to 7 GPa (in accordance with experimental values of Young modulus  $\sim 3 - 20$  GPa [7]) is modeled in the framework of Landau theory. The dependencies of ordinary magnetostriction on the temperature and stress are computed to promote the experimental studies and to prevent the misunderstanding of their results.

### Theoretical Grounds

**Martensitic Transformation of Stressed Ni–Mn–Ga Single Crystal.** The MT of Ni–Mn–Ga alloy may be described using the following form [12,13] of Landau expansion for Gibbs potential:

$$G = F_m + c_2(u_2^2 + u_3^2)/2 + a_4u_3(u_3^2 - 3u_2^2)/3 + b_4(u_2^2 + u_3^2)^2/4 - (\sigma_2^{(eff)}u_2 + \sigma_3^{(eff)}u_3)/6, \quad (1)$$

where  $F_m$  is the Helmholtz free energy of magnetic subsystem of the crystal,  $c_2$ ,  $a_4$ , and  $b_4$  are the linear combinations of second- third- and forth-order elastic modules enumerated after [12],

$$u_2 = \sqrt{3}(\varepsilon_{xx} - \varepsilon_{yy}), \quad u_3 = 2\varepsilon_{zz} - \varepsilon_{yy} - \varepsilon_{xx}, \quad (2)$$

$$\sigma_2^{(eff)} = \sqrt{3}(\sigma_{xx}^{(eff)} - \sigma_{yy}^{(eff)}), \quad \sigma_3^{(eff)} = 2\sigma_{zz}^{(eff)} - \sigma_{yy}^{(eff)} - \sigma_{xx}^{(eff)},$$

$\varepsilon_{ik}$  are the strain tensor components in the coordinate frame joined to  $\langle 100 \rangle$  crystallographic directions,  $\sigma_{ik}^{(eff)}$  are the effective stresses induced by the axial compression and magnetic ordering of the crystal, i.e.  $\sigma_{\alpha}^{(eff)} = \sigma_{\alpha} + \sigma_{\alpha}^{(me)}$ , where  $\sigma_{\alpha}^{(me)}$  are the magnetoelastic stresses [13], which are related to the dimensionless magnetoelastic constant  $\delta$  and magnetization vector components  $M_i \equiv M(T)m_i$  in a following way:

$$\sigma_2^{(me)} = 6\sqrt{3}\delta M^2(T)(m_x^2 - m_y^2), \quad \sigma_3^{(me)} = 6\delta M^2(T)(2m_z^2 - m_y^2 - m_x^2). \quad (3)$$

The equilibrium states of the elastic single crystal can be determined from the minimum conditions for Gibbs potential Eq. 1. For the sake of certainty let us consider the phase transition from parent cubic phase of Ni–Mn–Ga to z-variant of tetragonal phase, i.e. the contraction of cubic unit cell in [001] direction. When the alloy specimen is stressed in [001] or [110] direction the value  $\sigma_2^{(eff)}$  is equal to zero and so,  $u_2 = 0$ . In this case the condition  $\partial G / \partial u_3 = 0$  results in equation

$$u_3[c_2(T) + a_4u_3 + b_4u_3^2] - \sigma_3^{(eff)}(T, \sigma)/6 = 0. \quad (4)$$

In the case of moderate loading the solution of Eq. 4 can be approximated by superposition of the MT strain  $u_3^{(0)}(T) = 2[c(T)/a(T) - 1]$ , which arise on cooling of the unstressed alloy, and the elastic strain  $\tilde{u}(T, \sigma)$ , that is caused by the effective stress, i.e.  $u_3(T, \sigma) = u_3^{(0)}(T) + \tilde{u}(T, \sigma)$ . The MT strain corresponds to zero stress value and is expressed from Eq. 4 as

$$u_3^{(0)}(T) = \begin{cases} -(a_4/2b_4)(1 + \sqrt{1 - c_2(T)/c_t}) & \text{if } T < T_1 \\ 0 & \text{otherwise.} \end{cases} \quad (5)$$

The temperature  $T_1$  is the lability temperature of unstressed paramagnetic tetragonal phase, it satisfies condition  $c_2(T_1) = c_t \equiv a_4^2/4b_4 > 0$ . The lability temperature of unstressed paramagnetic cubic phase  $T_2$  satisfies the equation  $c_2(T_2) = 0$ . As long as the inequality  $|\tilde{u}(T, \sigma)| \ll |u_0(T)|$  is hold, the elastic strain satisfies the equation

$$\tilde{u}(T, \sigma) = -A^{-1}(T) \left\{ C_{pm}(T) - [C_{pm}^2(T) + A(T)\sigma_3^{(eff)}(T, \sigma)]^{1/2} \right\}, \quad (6)$$

where

$$C_{pm}(T) = 3c_2(T) + 6a_4u_3^{(0)}(T) + 9b_4[u_3^{(0)}(T)]^2, \quad A(T) = 6[a_4 + 3b_4u_3^{(0)}(T)]. \quad (7)$$

The equation (6) is valid if  $C_{pm}^2(T) > A(T)|\sigma_3^{(eff)}(T, \sigma)|$ , for more details see [14].

**Shear Modulus and Magnetostriction.** The function  $C_{pm}(T)$  describes the temperature dependence of shear elastic modulus of the crystal in the unstressed paramagnetic state. The shear modulus of deformed ferromagnetic crystal is defined as

$$C_{fm}(T, \sigma) = -(d\tilde{u} / d\sigma)^{-1}. \quad (8)$$

The equations (6)–(8) are sufficient for computation of shear modulus if the coefficients of Landau expansion for Gibbs potential, which are involved in Eq. 1, and the temperature dependencies of lattice parameters, which predetermine the  $u_3^{(0)}(T)$  function, are known.

To retard the transformation of  $z$ -variant of tetragonal lattice into  $y$ - and  $x$ -variants, it is convenient to apply magnetic field in  $[110]$  direction. In this case the transversal (with respect to the magnetic field direction) magnetostriction of martensite can be evaluated from the equation

$$\varepsilon_{zz}^{(ms)}(T, H, \sigma) \approx \frac{1}{3}[\tilde{u}(T, H, \sigma) - \tilde{u}(T, 0, \sigma)], \quad (9)$$

which result from the Eq. 2 and tendency to volume conservation during MT ( $\varepsilon_{zz} \approx -2(\varepsilon_{xx} + \varepsilon_{yy})$ ).

## Computations

**Input Data.** All computations were carried out using the fixed value of dimensionless magnetoelastic constant  $\delta = -23$  substantiated in [1] and articles cited therein. According to experimental data [7,8] the minimal value of shear modulus  $C_{fm}(T_1, 0) = 1$  GPa was accepted. This value together with experimental values of lattice distortion (measured for  $\text{Ni}_{1.99}\text{Mn}_{1.14}\text{Ga}_{0.87}$  alloy [15]), Curie temperature  $T_C$ , and saturation magnetization enabled determination of the parameters  $a_4 = 11$  GPa,  $b_4 = 92$  GPa [14] and computation of the functions  $u_3^{(0)}(T)$  and  $M(T)$ . The function  $c_2(T)$  was computed than from the Eqs. 4, 5. The computed functions are presented in Fig. 1. The computational procedure is explained in [14].

**Results.** The theoretical temperature dependence of magnetostriction of Ni–Mn–Ga single crystal, which undergoes phase transition from cubic to single-variant tetragonal state under the saturating magnetic field and weak axial compression, is shown in Fig. 2. The temperature dependence of shear modulus is shown in the Inset. The figure 2 illustrates the features of magnetostriction of ultra-soft FSMA-s belonging to Ni–Mn–Ga alloy family.

i) The magnetostriction  $\varepsilon_{zz}^{(ms)}(T, H_s, \sigma)$  is positive and almost constant well below MT temperature, its value ( $\sim 0.03\%$ ) is sufficient for technical applications. The constancy of this value has a fundamental reason: the magnetostriction is directly proportional to magnetoelastic stress  $\sigma^{(me)} \sim \delta M^2(T)$  (see Eq. 3) and inversely proportional to the elastic modulus of alloy specimen. Well below MT both magnetization and elastic modulus are the smoothly decreasing functions of temperature and their temperature dependencies compensate each other.

ii) The magnetostriction reaches a peak value in the vicinity of MT due to the abrupt softening of shear modulus. The magnetostriction curve is not symmetric with respect to the peak. This feature

retards the interpretation of experimental results. In particular, the energies of magnetoelastic coupling in martensitic and austenitic phases cannot be compared using the values of magnetostriction measured for two temperatures only. This statement is substantiated in Fig. 2. On the one hand, the magnetostriction of austenite read at 304 K is the double of magnetostriction of martensite read at 285 K (see points *A* and dashed arrows in Fig. 2). On the other hand, the magnetostriction of martensite read at 294 K is the double of magnetostriction of austenite read at 317 K (see points *B* and solid arrows in Fig. 2). However, all values of magnetostriction result from the same value of magnetoelastic constant  $\delta$ .

iii) The magnetostriction of austenite becomes negligibly small on approach to Curie temperature due to the reduction of magnetization value.

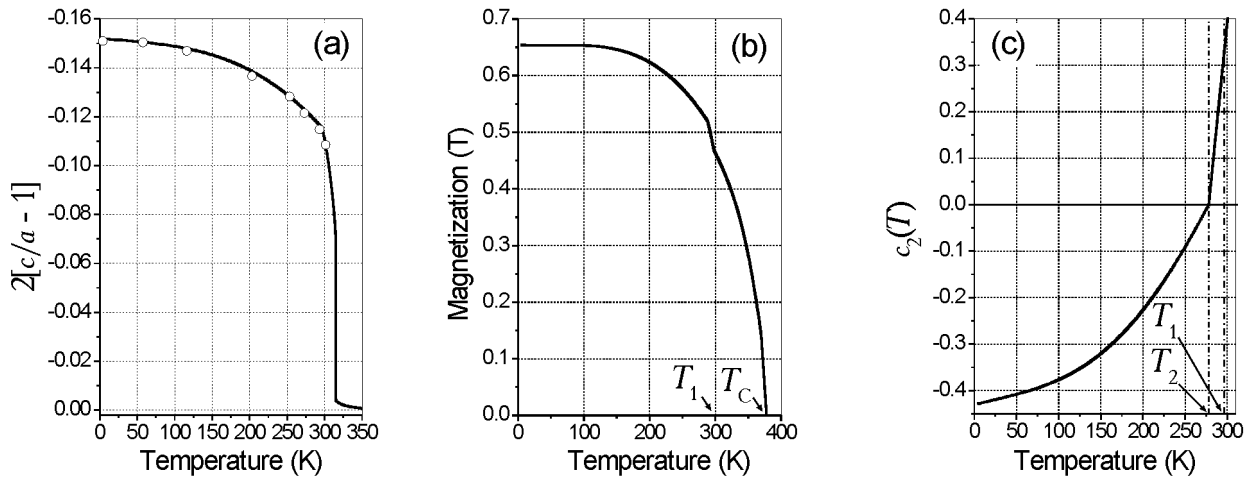


Fig. 1. Temperature dependencies of lattice distortion, (a), magnetization, (b), and coefficient  $c_2$  of Landau expansion for Gibbs potential, (c).

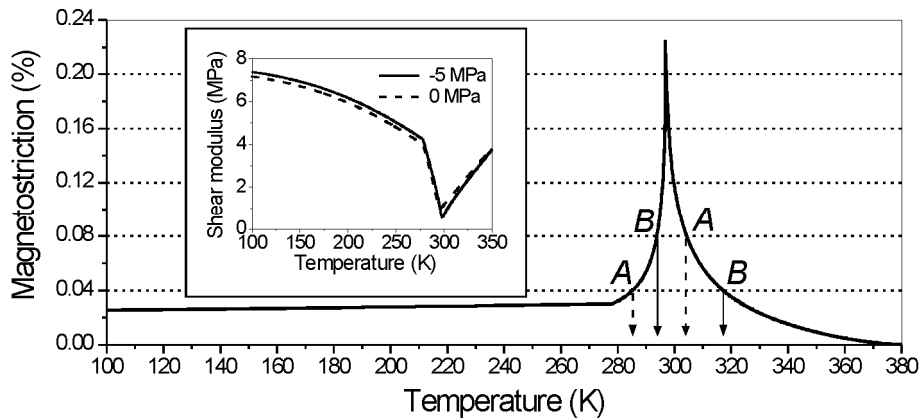


Fig. 2. Temperature dependencies of saturation magnetostriction of Ni–Mn–Ga single crystal compressed with  $-5$  MPa stress. The temperature dependencies of shear elastic modulus computed for zero and  $-0.5$  MPa stress values are shown in the Inset. The marks "A" and "B" are explained in the text.

The theoretical evaluations of elastic modules and magnetostriction of stressed Ni–Mn–Ga alloy are of practical and academic interest, because the reported experimental values are very different and disputed at present (see [3–9] and references therein). In particular, the magnetically induced

deformation of axially compressed specimen was measured and the reduction of deformation by factor  $\sim 0.3$  on stress variation from  $-15$  MPa to  $-60$  MPa was observed.<sup>9</sup> This reduction was conceivably attributed to the transformation of residual fragments of martensite structure "surviving" under the  $-60$  MPa stress. The theoretical analysis shows, however, that the significant elevation of shear modulus and the appropriate reduction of ordinary magnetostriction are possible even if the alloy is in the single variant state.

The influence of axial stress on the saturation magnetostriction and shear elastic modulus of single crystalline specimen of ultra-soft Ni–Mn–Ga alloy is illustrated in Fig. 3.

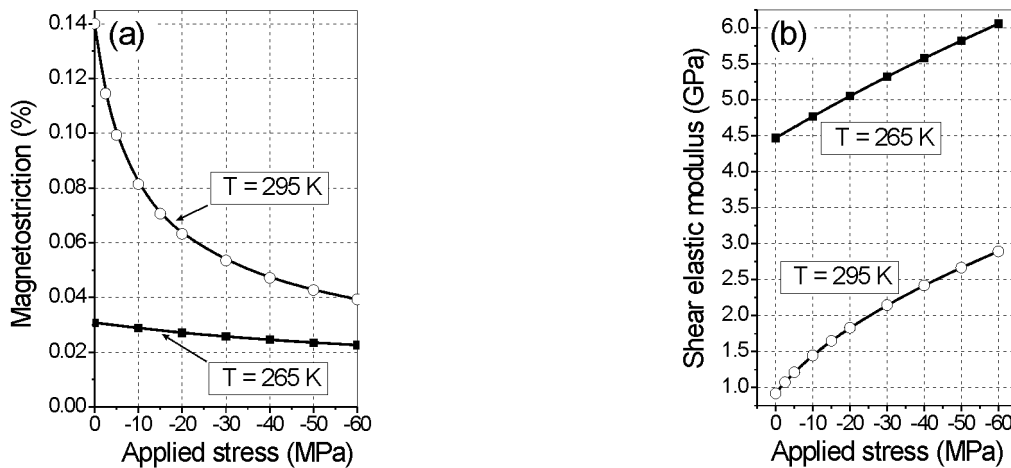


Fig. 3. The influence of axial compression on the saturation magnetostriction, (a), and shear elastic modulus, (b), of the single-variant martensite.

As it is seen from the figure, the affect of stress on the elastic and magnetoelastic behavior of the single crystalline specimen of ultra-soft alloy can be observed in the wide temperature range, but is the most pronounced in the vicinity of MT temperature. The computations showed that the application of  $-60$  MPa stress results in the diminution of magnetostriction by factor of 1.5, when the temperature of alloy is equal to 265 K, and by factor of 3.5 at room temperature (see Fig. 3 (a)). The shear elastic modulus of compressed single crystal obviously correlates to magnetostriction and demonstrates the inverse proportionality to it (see Fig. 3 (b)).

## Discussion

Let the Ni–Mn–Ga martensite is brought into the quasi-single-variant state with dominating  $z$ -variant of crystal lattice. It is commonly recognized now that in this case the giant deformation in the magnetic field aligned with  $[100]$  direction is caused by transformation of  $z$ -variant of the lattice into  $x$ -variant. If the magnetic field is applied in  $[110]$  direction this transformation is hindered, and therefore, this field direction and the proper orientation of single-variant specimen may be preferable for experimental determination of the ordinary magnetostriction of martensitic phase.

Another step, which may promote the correct determination of ordinary magnetostriction of martensitic phase, is the cooling of Ni–Mn–Ga single crystal subjected to the moderate compression. However, the stress exceeding the value of 10 MPa can substantially reduce magnetostriction and the reduction must be taken into account for the correct interpretation of experimental data. The quantitative account of stress influence needs, in its turn, the knowledge of temperature dependence of shear elastic modulus. The consideration of this dependence is absolutely necessary for the evaluation of the energy of magnetoelastic coupling from the magnetostriction measurements.

The study of ordinary magnetostriction of Ni–Mn–Ga alloys is not only of academic interest but of practical importance as well. The magnetostriction becomes almost independent on the temperature when the temperature dependencies of shear elastic modulus and magnetization partially compensate each other. In this case the realistic values of shear elastic modulus result in magnetostriction of about  $10^{-4}$ . It exceeds the magnetostriction of some magnetostrictive materials, which found practical application.

### Acknowledgements

This work is done in the frame of the partner project P-279 (EOARD 68008) with the funding support of European Office of Aerospace Research & Development (EOARD), USA. One of the authors (V. A. L.) would like to thank Dr. Aleksiy Sozinov and Dr. Volodymyr Chernenko for fruitful discussion.

### References

- [1] V.A. Chernenko and V.A. L'vov: Mater. Sci. Forum Vol. 583 (2008), p. 1.
- [2] S.J. Murray, M. Marioni, S.M. Allen, R.C. O'Handley and T. A. Lograsso: Appl. Phys. Lett. Vol. 77 (2000), p. 886.
- [3] A.N. Vasil'ev, S.A. Klestov, R.Z. Levitin, V.V. Snegirev, V.V. Kokorin and V. A. Chernenko: Zh. Eksp. Teor. Fiz. Vol. 109 (1996), p. 973.
- [4] R. Tickle and R.D. James: JMMM Vol. 195 (1999), p. 627.
- [5] V.V. Kokorin and M. Wuttig: JMMM Vol. 234 (2001), p. 25.
- [6] O. Heczko: JMMM Vol. 290 (2005), p. 846.
- [7] V.A. Chernenko, J. Pons, C. Segun and E. Cesari: Acta Mater. Vol. 50 (2002), p. 53.
- [8] L. Dai, J. Cullen and M. Wuttig: J. Appl. Phys. Vol. 95 (2004), p. 6957.
- [9] A. Gonzalez-Comas, E. Obrado, Ll. Manosa, A. Planes, V.A. Chernenko, B.J. Hattink and A. Labarta: Phys. Rev. B Vol. 60 (1999), p. 7085.
- [10] V.A. Chernenko and V.A. L'vov: Phil. Mag. A Vol. 73 (1996), p. 999.
- [11] O. Heczko, N. Glavatska, V. Gavriluk and K. Ullakko: Mater. Sci. Forum Vol. 373–376 (2001), p. 341.
- [12] V.A. Chernenko, O. Babii, V.A. L'vov, P.G. McCormick: Mater. Sci. Forum Vol. 327–328 (2000), p. 485.
- [13] V.A. Chernenko, V.A. L'vov, E. Cesari and P. McCormick: Mat. Trans. JIM Vol. 8 (2000), p. 928.
- [14] V.A. L'vov, N. Glavatska, I. Aaltio, O. Söderberg, I. Glavatskyy and S-P. Hannula: accepted to Acta Materialia (2009).
- [15] N. Glavatska, I. Glavatskiy, G. Mogilny, S. Danilkin, D. Hohlwain, O. Söderberg, V. Lindroos and A. Beskrovniy: J. Phys. IV (France) Vol. 112 (2003), p. 963.